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Temperature-Dependent Magnetoelectric Effect from First Principles

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We show that nonrelativistic exchange interactions and spin fluctuations can give rise to a linear magnetoelectric effect in collinear antiferromagnets at elevated temperatures that can exceed relativistic magnetoelectric responses by more than 1 order of magnitude. We show how symmetry arguments, *ab initio* methods, and Monte Carlo simulations can be combined to calculate temperature-dependent magnetoelectric susceptibilities entirely from first principles. The application of our method to Cr_2O_3 gives quantitative agreement with experiment.

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Introduction.—Recent years have seen a resurgence of interest in materials with coupled electric and magnetic dipoles motivated by the prospect of controlling spins with applied voltages and charges with applied magnetic fields in novel multifunctional devices. The simplest form of such a control is the linear magnetoelectric (ME) coupling between electric polarization and an applied magnetic field or, conversely, between magnetization and an applied electric field. Although the linear ME effect was theoretically predicted and experimentally discovered more than 50 years ago [1], finding technologically useful materials displaying strong ME coupling at room temperature remains a challenging problem [2].

Recent progress in the related field of multiferroic materials, in which ferroelectric polarizations are induced by noncentrosymmetric magnetic orderings, has led to a clarification of the microscopic origins for ME coupling [3]. In particular, two distinct coupling mechanisms have been identified. The first arises from relativistic effects linking electron spin and orbital momentum, resulting in the antisymmetric $\mathbf{S}_1 \times \mathbf{S}_2$ interaction between spins of different magnetic ions. The strength of this Dzyaloshinskii-Moriya interaction depends on polar displacements of ions, which can make magnets with noncollinear spiral orders becoming ferroelectric [4–7]. In the second mechanism, polar deformations of the lattice are induced by Heisenberg spin exchange interactions $\mathbf{S}_1 \cdot \mathbf{S}_2$, originating from the Fermi statistics of electrons [8,9]. This nonrelativistic mechanism can give rise to stronger spin-lattice couplings than those resulting from relativistic effects, which tend to be relatively weak in 3D transition metal compounds. Indeed, in multiferroics, the electric polarizations induced by exchange interactions in $\text{Y}_{1-x}\text{Lu}_x\text{MnO}_3$ and GdFeO_3 exceed the largest polarizations observed in spiral multiferroics by 1 order of magnitude [10,11]. It was recently suggested that Heisenberg exchange can also give rise to a relatively strong linear ME effect [12] which, however, seemed to require rather special noncollinear spin orderings and crystal structures, making it difficult to find such materials in nature.

In this Letter, we show that the Heisenberg exchange mechanism also works in antiferromagnets with simple collinear orderings. It gives rise to a ME effect that reaches its maximum at elevated temperatures where spin fluctuations are large. As an example, we use Cr_2O_3 —the first material in which the linear ME effect was discovered. By computing the temperature-dependent ME susceptibility of this material entirely from first principles, we show that its strong temperature dependence originates from the exchange mechanism.

This is not a straightforward computation since the ME response driven by Heisenberg interactions, which is strong at elevated temperatures, completely vanishes at zero temperature where *ab initio* methods apply. To circumvent this problem we extract the relevant ME couplings by calculating the electric polarizations of artificially imposed multiferroic spin orderings at zero temperature. The magnetoelectric susceptibility is then expressed in terms of spin correlation functions, which are calculated by using Monte Carlo simulations of the magnetic state at various temperatures. We show that spin fluctuations play a crucial role in the ME response of collinear magnets.

This work extends first-principles studies of magnetoelectric materials beyond their current boundaries and provides a unified approach to seemingly different phenomena, such as the linear ME effect and multiferroicity. It clarifies the dominant mechanism of the ME coupling in Cr_2O_3 and accurately describes the temperature dependence of its response with no adjustable parameters. Furthermore, since collinear antiferromagnets with high ordering temperatures are not uncommon, our study opens a route to engineering strong ME responses at room temperature.

Origin of strong magnetoelectric coupling in Cr_2O_3 .—The ME effect in Cr_2O_3 was predicted phenomenologically by Dzyaloshinskii [1] and measured by Astrov [13] shortly after the theoretical prediction. Cr_2O_3 has four magnetic Cr^{3+} ions in the rhombohedral unit cell. Below $T_N = 307$ K, it develops a collinear $\uparrow\downarrow\uparrow\downarrow$ spin ordering along the trigonal z axis [14] [see Fig. 1(a)]. This ordering

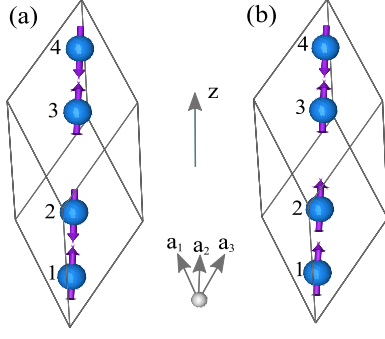


FIG. 1 (color online). Rhombohedral unit cell of Cr_2O_3 with the unit vectors \mathbf{a}_i ($i = 1, 2, 3$) containing four magnetic Cr ions. (a) shows the actual antiferromagnetic spin ordering in Cr_2O_3 , while (b) shows the spin ordering imposed in our first-principles calculations to induce an electric polarization along the trigonal z axis.

breaks the inversion symmetry of the paramagnetic phase, reducing it to inversion combined with time reversal, allowing for two independent ME coupling terms in the free energy [1]:

$$F_{\text{me}} = -\alpha_{\parallel} E^z H^z - \alpha_{\perp} (E^x H^x + E^y H^y), \quad (1)$$

where the ME coefficients α_{\parallel} and α_{\perp} are proportional to the antiferromagnetic order parameter $G^z = \langle S_1^z - S_2^z + S_3^z - S_4^z \rangle$. These two ME coefficients show very different temperature dependences [13,15]. While α_{\perp} is temperature-independent up to $T \sim T_N$ and relatively small, the ME response described by α_{\parallel} increases dramatically from close to zero at low temperature, reaching its maximum at $T_{\text{max}} \sim 260$ K. At its peak, the magnitude of α_{\parallel} is 1 order of magnitude larger than $|\alpha_{\perp}|$ at any T and $\alpha_{\parallel}(T = 0)$.

Early measurements of the temperature dependence of the ME coefficients [16–18], as well as recent first-principles calculations [19], have shown that the relatively weak ME effects at low temperature result from relativistic interactions. In what follows, we demonstrate that the much stronger response at elevated temperatures originates from Heisenberg exchange.

Phenomenologically, the electric polarization P^z along the trigonal axis can couple to spins as follows:

$$P^z = \lambda (\mathbf{S}_1 \cdot \mathbf{S}_3 - \mathbf{S}_2 \cdot \mathbf{S}_4), \quad (2)$$

where \mathbf{S}_1 – \mathbf{S}_4 denote the magnetization of sites 1–4, respectively, and λ is the coupling strength that we will determine from *ab initio* calculations. Here we have taken into account the fact that the exchange-driven polarization can depend only on scalar products of the magnetizations. The combination of scalar products in the right-hand side of Eq. (2) transforms in the same way as P^z ; this can be seen by inspection of Table I, which shows how the four inequivalent magnetic sites transform under the symmetry operations of Cr_2O_3 .

Equation (2) is clearly appropriate for describing electric polarization induced by spin ordering in a multiferroic

TABLE I. Transformation of four independent Cr sites with the fractional coordinates $\mathbf{r}_1 = (u, u, u)$, $\mathbf{r}_2 = (1/2 - u, 1/2 - u, 1/2 - u)$, $\mathbf{r}_3 = (1/2 + u, 1/2 + u, 1/2 + u)$, and $\mathbf{r}_4 = (1 - u, 1 - u, 1 - u)$, where $u \approx 0.153$, under the generators of space group $R\bar{3}c$: the 120° rotation around the z axis, $C_3 = (x_3, x_1, x_2)$, the 180° rotation around the axis orthogonal to the z direction, $C_2 = (1/2 - x_2, 1/2 - x_1, 1/2 - x_3)$, and inversion $I = (1 - x_1, 1 - x_2, 1 - x_3)$. Here, $\mathbf{c} = \mathbf{a}_1 + \mathbf{a}_2 + \mathbf{a}_3$, where \mathbf{a}_i ($i = 1, 2, 3$) are the rhombohedral unit vectors.

	C_3	C_2	I
1	1	2	4
2	2	1	3
3	3	4 - c	2
4	4	3 - c	1

material. In addition, it applies to the linear ME effect, as can be seen from the following heuristic argument: In an applied magnetic field H^z the average value of spin on the sublattice α changes by $\langle \delta S_{\alpha}^z \rangle \propto \chi_{\parallel} H^z$, where χ_{\parallel} is the longitudinal magnetic susceptibility. Equation (2) then gives $P^z \propto \lambda \chi_{\parallel} \langle S_1^z - S_2^z + S_3^z - S_4^z \rangle H^z \propto \lambda \chi_{\parallel} G^z H^z$, consistent with $P^z = -\frac{\partial F_{\text{me}}}{\partial E^z} = \alpha_{\parallel} H^z$ obtained from Eq. (1).

Equation (2) is meaningful only within the mean-field approach. To account for effects of spin fluctuations on the ME response of Cr_2O_3 , we will use the microscopic expression for the exchange-driven polarization in terms of scalar products of Cr spins (rather than the sublattice magnetizations), which has the form

$$P^z = \frac{\lambda}{6N} \sum_j \sum_{n=1}^6 (\mathbf{S}_{1,j} \cdot \mathbf{S}_{3,j-b_n} - \mathbf{S}_{2,j} \cdot \mathbf{S}_{4,j-b_n}). \quad (3)$$

Here j labels unit cells, N is the total number of unit cells, $\mathbf{b}_1 = \mathbf{a}_1$, $\mathbf{b}_2 = \mathbf{a}_2$, $\mathbf{b}_3 = \mathbf{a}_3$, $\mathbf{b}_4 = \mathbf{a}_1 + \mathbf{a}_2$, $\mathbf{b}_5 = \mathbf{a}_2 + \mathbf{a}_3$, and $\mathbf{b}_6 = \mathbf{a}_3 + \mathbf{a}_1$ (\mathbf{a}_i being the rhombohedral unit vectors). Remarkably, the exchange interaction between the fourth nearest-neighbor Cr ions separated by the distance $r_{1,(3-\mathbf{a}_1)} = r_{1,(3-\mathbf{a}_1-\mathbf{a}_2)} = 3.65 \text{ \AA}$ turns out to give rise to the linear ME effect in Cr_2O_3 . The shorter-range exchange interactions do not couple the sublattices 1 and 3 (or 2 and 4) and, therefore, do not contribute to P^z , while other interactions between these sublattices correspond to much longer exchange paths and are negligibly small. This allows us to accurately pinpoint the microscopic origin of the strong ME effect in Cr_2O_3 .

Importantly, Eqs. (2) and (3) apply to any four-sublattice spin ordering in Cr_2O_3 . The $\uparrow\downarrow\uparrow\downarrow$ spin ordering realized in the low-temperature ground state of Cr_2O_3 induces no electric polarization, and so a straightforward density-functional study of Cr_2O_3 does not give information about α_{\parallel} at finite temperatures. However, the $\uparrow\uparrow\uparrow\downarrow$ ordering shown in Fig. 1(b) renders Cr_2O_3 multiferroic and induces the electric polarization

$$\mathcal{P} \equiv P^z(\uparrow\uparrow\uparrow\downarrow) = 2\lambda S^2. \quad (4)$$

We next extract λ by using *ab initio* methods by enforcing

ing $\uparrow\uparrow\uparrow$ spin ordering and calculating the magnetically induced polarization \mathcal{P} .

First-principles calculations of the magnetoelectric coupling.—We compute λ and the spin exchange parameters by using plane-wave density-functional theory, as implemented in the Vienna *ab initio* simulation package (VASP) [20]. We use the projector-augmented wave method [21] for core-valence partitioning and the local-spin-density approximation with a rotationally invariant Hubbard- U for the exchange-correlation potential [22]. Our Hubbard $U = 2.0$ eV is the same value that was taken for computing the perpendicular magnetoelectric response at zero temperature [19]. In order to demonstrate the nature of \mathcal{P} , we deliberately do not include spin-orbit coupling, so that λ corresponds only to polarizations induced by exchange mechanism.

We work with space group $R\bar{3}c$ at the experimental volume [23] of 96.0 \AA^3 and rhombohedral angle of 55.13° . The internal coordinates are relaxed within our density-functional calculations for the $\uparrow\downarrow\uparrow$ magnetic configuration, yielding coordinates $x = 0.1536$ for Cr and $x = 0.9426$ for O in Wyckoff positions $4c$ and $6e$, respectively. Subsequently, the Heisenberg exchange couplings J_1 - J_5 [24], corresponding to Cr-Cr distances of 2.65 - 4.10 \AA , are computed by fitting a Heisenberg Hamiltonian to density-functional theory total energies of 12 different spin configurations with fixed ion coordinates in the hexagonal setting of $R\bar{3}c$. This method is analogous to that employed by Shi, Wysocki, and Belashchenko [25].

Finally, we compute λ by enforcing the spin configuration of $\uparrow\uparrow\uparrow$ and relaxing again the ionic coordinates in the rhombohedral unit cell. The resulting ionic configuration has a polar lattice distortion. We compute the magnitude of $\mathcal{P} = 0.585 \text{ } \mu\text{C}/\text{cm}^2$ by using the Berry phase approach [26], which allows us to extract λ . We note that \mathcal{P} is of the same order of magnitude as the polarization induced by exchange interactions in multiferroics with collinear spins [10,11].

Monte Carlo simulations.—Using Eq. (3), we can now express the temperature-dependent ME coefficient α_{\parallel} in terms of spin correlation functions:

$$\alpha_{\parallel} = \left. \frac{\partial \langle P^z \rangle}{\partial H^z} \right|_{H^z=0} = \frac{2\mu_B}{k_B T} \left\langle P^z \sum_{\alpha,j} S_{\alpha,j}^z \right\rangle, \quad (5)$$

where $\langle \dots \rangle$ denotes the thermal average at temperature T , k_B is the Boltzmann constant, and μ_B is the Bohr magneton.

In the mean-field approximation (details in supplementary material [27]) one obtains [16,17]

$$\alpha_{\parallel} = \frac{\lambda v_0 G^z \chi_{\parallel}}{8\mu_B}, \quad (6)$$

where v_0 is the unit cell volume, in agreement with the simple argument given above. The mean-field expression qualitatively explains the observed temperature dependence of α_{\parallel} : It first grows, together with the order param-

eter G^z , as the temperature drops below T_N , and then subsequently decreases and vanishes at $T = 0$, together with the longitudinal magnetic susceptibility χ_{\parallel} .

In Ref. [18] an attempt was made to take into account the effects of spin fluctuations by using a higher-order decoupling scheme. This approximation fails, however, close to the transition temperature where spin fluctuations are large. We include spin fluctuations by calculating α_{\parallel} numerically by using Monte Carlo simulations of a system of 864 classical spins with exchange constants and the magnetoelectric coupling λ obtained from our first-principles calculations, as described above.

In a finite-sized system of Heisenberg spins, the antiferromagnetic order parameter is free to rotate during the simulation, which corresponds to exploration of different domains. The correlation function on the right-hand side of Eq. (5) is zero unless a single antiferromagnetic domain is selected. We therefore apply to our system a weak staggered field along the z axis, $h(-)^{\alpha}$, where $\alpha = 1, 2, 3, 4$ labels magnetic sublattices. This field mimics the presence of the magnetic easy axis of Cr_2O_3 as well as the magnetoelectric annealing used in experiment to select the domain with a given sign of magnetic order parameter. The field strength was chosen so that it is small compared to the scale of exchange interactions but large enough to make the Monte Carlo results independent of h .

Figure 2 shows the temperature dependence of α_{\parallel} obtained from Monte Carlo simulations (blue circles) and in the mean-field approximation described above (red solid line). The onset of the ME response in our Monte Carlo simulations, as well as the sharp peak in the specific heat (inset), shows that the antiferromagnetic order sets in at $\sim 290 \text{ K}$, close to the experimentally observed transition

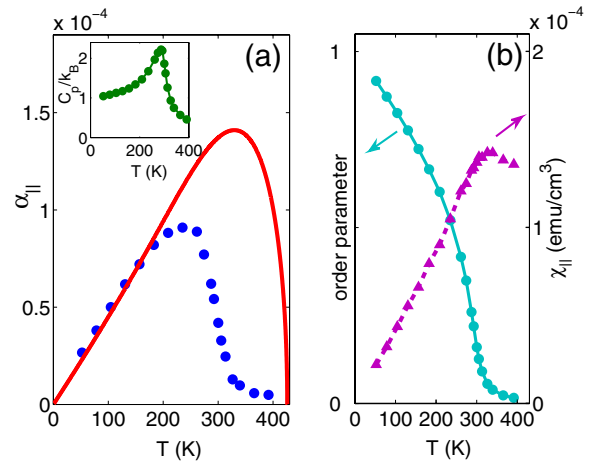


FIG. 2 (color online). (a) Temperature dependence of the magnetoelectric coupling α_{\parallel} obtained by using *ab initio* values of the exchange constants and magnetoelectric coupling combined with Monte Carlo simulations (blue circles) and mean-field calculations [27] (solid red line). The inset shows the temperature dependence of magnetic specific heat. The green line is a guide to the eye. (b) Evolution of the antiferromagnetic order parameter and the magnetic susceptibility with temperature.

temperature $T_N = 307$ K. The maximal value of the ME coefficient obtained from our simulations is 0.9×10^{-4} (in Gaussian units), in excellent agreement with the experimental value of 1.0×10^{-4} (see Ref. [15]). The maximum value is reached at ~ 240 K that compares well to $T_{\max} \sim 260$ K found in experiment. The mean-field transition temperature (425 K) and maximal α_{\parallel} are significantly higher than the Monte Carlo values, indicating the importance of spin fluctuations in this material.

Conclusions.—We have presented the first *ab initio* calculation of a temperature-dependent linear magnetoelectric response. The quantitative agreement of our results with experimental data on Cr_2O_3 demonstrates that the dominant magnetoelectric coupling in this material originates from nonrelativistic exchange interactions between electrons. The strong temperature dependence of the magnetoelectric coefficient α_{\parallel} underscores the general importance of spin fluctuations for magnetoelectric responses of materials with collinear spin orders. Our study shows that the linear magnetoelectric effect and multiferroicity originate from the same microscopic mechanisms and can be described in a unified way. In both cases magnetoelectric effects resulting from nonrelativistic exchange interactions exceed relativistic effects by more than 1 order of magnitude.

Finite-temperature calculations using a similar approach have been reported previously for fixed temperature [28], but in this case the couplings could be extracted directly from first-principles calculations by using perturbations around the ground state. The approach used in this Letter, specifically the combination of first-principles calculations for artificially imposed magnetic states with Monte Carlo simulations of magnetoelectric response at elevated temperatures, opens a route to theoretical studies of a large variety of temperature-dependent static and dynamic magnetoelectric phenomena. It applies to other materials with strong temperature variations of ME susceptibility, such as Fe_2TeO_6 [29], $(\text{GaFe})\text{O}_3$ [30], and Ti_2O_3 [31], and can be extended to account for relativistic effects. Accurate predictions of the magnitude of magnetoelectric responses at finite temperature will greatly facilitate the search for and design of materials with the strongest responses.

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